Atmos. Chem. Phys. Discuss., 6, S683–S686, 2006
www.atmos-chem-phys.org/acpd/6/S683/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "An inverse modeling procedure to determine particle growth and nucleation rates from measured aerosol size distributions" *by* B. Verheggen and M. Mozurkewich

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Received and published: 28 April 2006

Both referees suggest that we should make the computer code available to aid other researchers in using this method. We agree with the spirit of this; however, it is somewhat problematic in that the program is not written as subroutines in a standard programming language and does not have a user interface. As a result, it is not straightforward for others to use "off the shelf". So we are concerned that by simply posting the existing code we will create a technical support problem. It is envisaged to make the program more user friendly by re-programming certain parts and by creating a user interface,

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after which the program (interface and code) will be made available via the internet. However, this constitutes a future project for which funding is currently sought. Of course, we are willing to share the program code if requested, provided that we have a clear understanding with prospective users as to what is expected in terms of support.

We thank the reviewer for pointing out a number errors and places where the text was unclear. We will make appropriate corrections and revisions. Replies on discussion items raised by Referee #1 are given below.

Page 1689, line 11. Coagulation with small particles. The referee suggests that we use the calculated concentration of particles below the minimum detectable size to recursively include coagulation with these very small particles in the regression analysis. This would require an iterative procedure, where the growth rate depends on the calculation backwards in time, and vice-versa. It certainly is an interesting suggestion, which we will consider when further developing this method. We do not believe that it is necessary at this stage since in most cases the growth due to within-mode coagulation of very small particles is small compared to the growth due to condensation.

Page 1698, line 10-12. The reviewer asks how the average growth rate, derived from the conventional banana contour plot, compares with the growth rates derived from the inverse modeling. The growth rate from fitting a curve through the banana contour plot has a similar average value as that deduced from the regression analysis, but it lacks the detailed time evolution. The following sentence will be added to the revised text (page 1698, line 9): "From inspection of the contour plot (Fig. 2) an average growth rate of 6 (+/- 1) nm h-1 can be determined for the time interval from 12:00 to 13:30, with a slight decrease in magnitude from beginning to end."

Section 4.2. The reviewer asks how the results for the wall loss and coagulation rate fit parameters, C_diff and C_coag, compare between experiments. The following sentence will be added to the revised text (page 1696, line 22): Since the wall loss and coagulation rates can only be accurately determined when the condensational growth

ACPD

6, S683–S686, 2006

rate is zero (or accurately known), not all experiments are equally suitable to determine C_diff and C_coag. However, similar values of these parameters were found from regression analysis of another experiment, suggesting that the wall loss and coagulation rates were relatively constant from experiment to experiment. This will be mentioned in the revised text.

Page 1708, line 7. Suggested comparison to the Vehkamäki et al. (2002) parametrization. The Vehkamäki et al. (2002) paper updates the previous parameterization and extends it to lower temperature and lower relative humidity, which do not apply to the measurements presented here. When we tried to use the newer parameterization to calculate the threshold concentration of H_2SO_4 we obtained a result of infinity for the conditions in these experiments. From the graph given by Vehkamäki et al., it appears that the difference in the two parameterizations is not large for our conditions. We therefore continue to use the Kulmala et al. (1998) reference.

The reviewer has suggested that in Figure 4 the backwards calculated size distributions should be extended to 0.5 nm radius. The data do not permit this for the case shown in the figure. The reconstructed size distributions are valid for 12:22 and are derived from measurements made at later times. None of those measurements provided reliable data on the size distribution below 1.4 nm. This is because of interference from the particles nucleated following the SO_2 injection at 12:50. As a result, no nucleation rate could be obtained for the time used for Figure 4. The primary purpose of this figure is to compare the various extrapolated distributions to each other and to the measured distribution. A secondary purpose of Figure 4 is to illustrate the concept of the nucleation rate calculation since those rates are determined from the concentration at 0.5 nm when they are available in the reconstructed distribution. Unfortunately, the text is misleading in this regard since it implies that a nucleation rate could actually be obtained from the results in Figure 4, rather than from similar figures plotted for other times. We will reword this.

If a similar figure is constructed for an earlier time, then the reconstructed distributions would extend to smaller sizes but there would be little or no overlap with the measured distributions since virtually all of the particles would be smaller than the smallest measured particles; such a figure would not serve to compare measured and reconstructed distributions. We have considered adding such a figure, but feel that it would add little to Figures 4 and 5.

The reviewer states that "It would be important to see the Figure extended to this size of 0.5 nm. It seems that the uncertainty in the concentration N_0.5 becomes very large for these small sizes. This uncertainty should be discussed." This is certainly an important point, but we felt it was served by Figure 5, which compares the nucleation rates; those are directly proportional to the concentrations at 0.5 nm.

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 1679, 2006.